

Temperature dependence of the normal-state Hall coefficient of a quasi-one-dimensional metal

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Abstract

We develop a systematic theory of the Hall effect in Q1D conductors in both weak and strong magnetic fields for a model where the electron relaxation time varies over the Fermi surface. At high temperatures, the Hall coefficient saturates at the value $-\beta/ecn$, where the dimensionless coefficient β is determined by the curvature of the longitudinal dispersion law of electrons, e the electron charge, c is the speed of light, and n is the hole concentration. At low temperatures, where a strong variation of the relaxation rate over the Fermi surface develops in the form of “hot spots”, the Hall coefficient becomes temperature-dependent and may change sign for a particular choice of the transverse dispersion law parameters. In our model, the sign changes in a weak, but not in a strong magnetic field.

Keywords: Many-body and quasiparticle theories; Transport measurements, conductivity, Hall effect, magneto-transport; Organic conductors based on radical cation and/or anion salts; Organic superconductors.

Recently, the **(a, b)**-plane Hall resistivity was measured under pressure in the normal state of $(\text{TMTSF})_2\text{PF}_6$ and was found to be strongly temperature-dependent and changing sign around 20 K [1]. We investigate a possible theoretical explanation of this effect within the “hot spots” model [2], where the electron relaxation time strongly varies over the Fermi surface. The often-used formula for the Hall coefficient $R_H = 1/enc$ [3] (where e and n are the electric charge and concentration of carriers, and c is the speed of light) applies only to the case of closed electron orbits in a strong magnetic field and does not apply to quasi-one-dimensional (Q1D) conductors, where the electron orbits are open. In this paper, we develop a systematic theory of the Hall effect in the latter case.

The $(\text{TMTSF})_2\text{X}$ materials can be modeled as layers of conducting chains parallel to the x axis and spaced at a distance b along the y direction. A magnetic field H is applied perpendicular to the layers. In this paper, we neglect coupling between the layers and study a two-dimensional (2D) problem. The results can be easily generalized to 3D by integrating over the momentum component perpendicular to the layers.

We start from the linearized stationary Boltzmann equation [3] written for the deviation $\chi(\mathbf{p})$ of the electron distribution function $f(\mathbf{p})$ from the equilibrium Fermi function $f_0(\varepsilon)$: $f(\mathbf{p}) - f_0(\varepsilon) = -\chi(\mathbf{p}) \partial f_0(\varepsilon) / \partial \varepsilon$, where \mathbf{p} and $\varepsilon(\mathbf{p})$ are the 2D electron momentum and

energy:

$$\frac{eH}{c} v(p_t) \frac{\partial \chi(p_t)}{\partial p_t} + \frac{\chi(p_t)}{\tau(p_t)} = e \mathbf{E} \cdot \mathbf{v}(p_t). \quad (1)$$

In Eq. (1), p_t is the component of the electron momentum tangential to the Fermi surface, which labels different points on the Fermi surface. $\mathbf{v}(\mathbf{p}) = \partial \varepsilon(\mathbf{p}) / \partial \mathbf{p}$ and $\tau(p_t)$ are the local values of the electron velocity and relaxation time at each point p_t on the Fermi surface. \mathbf{E} is an applied electric field, and e is the (negative) electron charge.

In the case of a weak magnetic field H , the first term in Eq. (1) is a small perturbation, so a solution can be obtained as a series in powers of H by iterating this term. The Hall conductivity per layer, σ_{xy} , is given by Ong’s formula [4]:

$$\sigma_{xy} = -\frac{2e^3 H}{(2\pi\hbar)^2 c} \oint dp_t v_y(p_t) \tau(p_t) \frac{d[v_x(p_t) \tau(p_t)]}{dp_t}, \quad (2)$$

where the integral over p_t is taken around the Fermi surface, and the coefficient 2 comes from the two spin orientations.

We apply the general formula (2) to a Q1D conductor, where the Fermi surface consists of two open sheets located in the vicinities of the the two longitudinal (along the chains) Fermi momenta $\pm p_F$. The electron dispersion laws in the vicinities of $\pm p_F$ can be written as

$$\varepsilon(\mathbf{p}) = \varepsilon_x(p_x) + \varepsilon_y(p_y), \quad (3)$$

$$\varepsilon_x = \pm v_F(p_x \mp p_F) - (p_x \mp p_F)^2/2m, \quad (4)$$

$$\varepsilon_y = 2t_b \cos(k_y \pm \varphi) + 2t'_b \cos(2k_y \pm \varphi'), \quad (5)$$

where the energy ε is measured from the Fermi energy, and $k_y = p_y b/\hbar$. The longitudinal energy dispersion law (4) is expanded to the second power of $p_x \mp p_F$. The coefficient of the first term is the Fermi velocity v_F , and the coefficient of the second term is

$$1/m = -\partial^2 \varepsilon_x / \partial p_x^2|_{p_F} = \beta v_F / p_F. \quad (6)$$

The longitudinal energy band in (TMTSF)₂X can be modeled as a tight-binding band 1/4-filled by holes or as a parabolic band for holes. In these cases, the dimensionless coefficient β in Eq. (6) has the following values:

$$\beta = \begin{cases} \pi/4 = 0.785 & \text{for } \varepsilon_x = 2t_a \cos(p_x a/\hbar), \\ 1 & \text{for } \varepsilon_x = -p_x^2/2m, \end{cases} \quad (7)$$

where t_a and a are the electron tunneling amplitude and spacing between the TMTSF molecules along the chains. In Eq. (5), t_b and t'_b are the electron tunneling amplitudes between the nearest and next-nearest chains, and the phases φ and φ' originate from the triclinic structure of the (TMTSF)₂X crystals [5]. (We otherwise ignore the triclinic structure, considering the x , y , and z axis to be mutually orthogonal.) Below, in Eqs. (8)–(14), we calculate the transport contribution from the $+p_F$ Fermi sheet and double the result to account for the $-p_F$ sheet.

A simple approximation where the electron relaxation time $\tau(p_t) = \tau$ and longitudinal velocity $v_x(p_t) = v_F$ are assumed to be constant over the Fermi surface is insufficient to calculate the Hall conductivity, because Eq. (2) gives $\sigma_{xy} = 0$ in this case. Thus, we need to take into account the second term $v_x^{(2)}$ in the expression for the longitudinal velocity obtained from Eq. (4) at the Fermi surface $\varepsilon(p_x, p_y) = 0$:

$$v_x = v_F + v_x^{(2)}, \quad v_x^{(2)} = -(p_x - p_F)/m \approx \varepsilon_y / m v_F. \quad (8)$$

Substituting the first term, v_F , into Eq. (2), integrating by parts, and averaging over k_y instead of p_t :

$$\int_0^{2\pi} \frac{dk_y}{2\pi} f(k_y) = \langle f(k_y) \rangle_{k_y}, \quad (9)$$

we find one contribution to the Hall conductivity:

$$\begin{aligned} \sigma_{xy}^{(1)} &= \frac{e^3 H v_F}{\pi \hbar^2 c} \left\langle \tau^2(k_y) \frac{\partial v_y(k_y)}{\partial k_y} \right\rangle_{k_y} \\ &= \frac{e^3 H v_F b}{\pi \hbar^3 c} \left\langle \tau^2(k_y) \frac{\partial^2 \varepsilon_y(k_y)}{\partial k_y^2} \right\rangle_{k_y}. \end{aligned} \quad (10)$$

Another contribution is obtained by substituting $v_x^{(2)}$ from Eq. (8) into Eq. (2):

$$\sigma_{xy}^{(2)} = -\frac{2e^3 H}{\pi \hbar c b m v_F} \langle \tau^2(k_y) v_y^2(k_y) \rangle_{k_y}$$

$$= -\beta \frac{2e^3 H b}{\pi \hbar^3 c p_F} \left\langle \tau^2(k_y) \left(\frac{\partial \varepsilon_y(k_y)}{\partial k_y} \right)^2 \right\rangle_{k_y}, \quad (11)$$

$$\sigma_{xy}^{(3)} = \beta \frac{e^3 H b}{2\pi \hbar^3 c p_F} \left\langle \tau^2(k_y) \frac{\partial^2 \varepsilon_y^2(k_y)}{\partial k_y^2} \right\rangle_{k_y}. \quad (12)$$

Eq. (11) originates from the derivative $\partial v_x^{(2)}(k_y)/\partial k_y$ in Eq. (2), and Eq. (12) comes from the derivative $\partial \tau(k_y)/\partial k_y$ integrated by parts.

For calculation of the diagonal components of the conductivity tensor, it is sufficient to use v_F for v and v_x :

$$\sigma_{xx} = \frac{2e^2 v_F}{\pi \hbar b} \langle \tau(k_y) \rangle_{k_y}, \quad (13)$$

$$\begin{aligned} \sigma_{yy} &= \frac{2e^2}{\pi \hbar b v_F} \langle \tau(k_y) v_y^2(k_y) \rangle_{k_y} \\ &= \frac{2e^2 b}{\pi \hbar^3 v_F} \left\langle \tau(k_y) \left(\frac{\partial \varepsilon_y(k_y)}{\partial k_y} \right)^2 \right\rangle_{k_y}. \end{aligned} \quad (14)$$

Using Eqs. (10)–(14), we calculate the Hall coefficient:

$$R_H = \sum_{i=1}^3 R_H^{(i)}, \quad \text{where} \quad R_H^{(i)} = \frac{\sigma_{xy}^{(i)}}{H \sigma_{xx} \sigma_{yy}}. \quad (15)$$

Of the three terms $R_H^{(i)}$ (10)–(12), only $R_H^{(2)}$ has a nonzero value $R_H^{(0)}$ when $\tau(k_y) = \text{const}$:

$$R_H^{(2)} \rightarrow R_H^{(0)} = -\beta / e c n, \quad (16)$$

where $n = 4p_F/2\pi \hbar b$ is the concentration of holes. The minus sign in Eq. (16) corresponds to the hole sign of the Hall effect. Eq. (16) differs from the conventional formula $R_H = 1/ecn$ by the coefficient β given by Eq. (7), which is proportional to the curvature (nonlinearity) of the longitudinal dispersion law (see Eq. (6)). Eq. (16) was obtained earlier in Ref. [6]. For a parabolic band, $\beta = 1$, and Eq. (16) coincides with the naive result. However, for a 1/4-filled tight-binding model, $R_H^{(0)}$ is reduced by the factor $\pi/4$. The curvature β and the Hall coefficient $R_H^{(0)}$ may be further reduced, if dimerization of the TMTSF stack is taken into account. For a half-filled tight-binding model, β and $R_H^{(0)}$ vanish.

On the other hand, $R_H^{(1)}$ and $R_H^{(3)}$ are completely determined by variation of the relaxation time $\tau(k_y)$ over the Fermi surface. $R_H^{(1)}$ also vanishes by symmetry when $t'_b = 0$ because of $\tau(k_y) = \tau(k_y + \pi)$. In Ref. [2], we calculated the distribution of the electron scattering rate $1/\tau(k_y)$ over the Fermi surface of a Q1D metal. We found that a strong variation of $1/\tau(k_y)$ develops at low temperatures, where the relaxation rate at certain “hot spots” becomes much higher than on the rest of the Fermi surface. In the present paper, following the same method, we compute the distribution

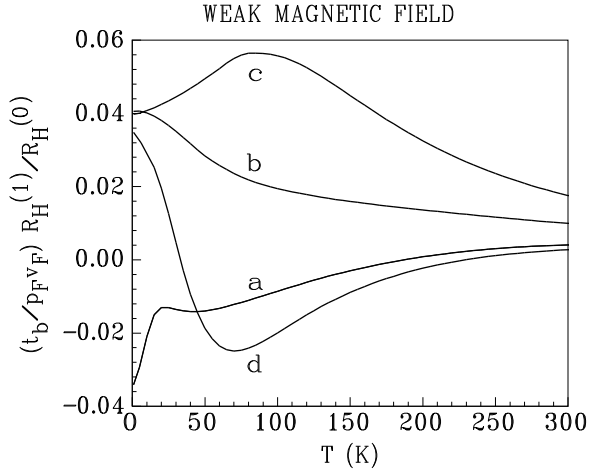


Figure 1: Temperature dependence of the first contribution $R_H^{(1)}$ to the Hall coefficient, normalized to $(v_F p_F / t_b) R_H^{(0)}$, in the case $\omega_c \tau \ll 1$.

of the electron relaxation rate $1/\tau(k_y)$ for the dispersion law (3)–(5) with $t_b = 300$ K, $t'_b = 30$ K, and several choices of φ and φ' . In Figs. 1–3, the curves (a) correspond to $\varphi = \varphi' = 0$, the curves (b) to $\varphi = \varphi'/2 = \pi/8$, the curves (c) to $\varphi = \varphi'/2 = \pi/4$, and the curves (d) to $\varphi = \pi/12$ and $\varphi' = 0$.

We substitute the computed $\tau(k_y)$ into Eqs. (10)–(15) and calculate $R_H^{(i)}$ as a function of temperature T . On one hand, $\sigma_{xy}^{(1)}$ (10) is relatively small, because it requires an asymmetry in the $\tau(k_y)$ distribution. On the other hand, it is proportional to the first power of the transverse tunneling amplitude t_b , whereas $\sigma_{xy}^{(2)}$ (11) is proportional to t_b^2 . Thus, $\sigma_{xy}^{(1)}$ is enhanced relative to $\sigma_{xy}^{(2)}$ by the big factor $v_F p_F / t_b$ equal to $\pi t_a / \sqrt{2} t_b$ in the 1/4 filled tight-binding model of (TMTSF)₂X. In Fig. (1), we show the temperature dependence of $R_H^{(1)}$ normalized to $(v_F p_F / t_b) R_H^{(0)}$ (with $\beta = \pi/4$). We observe that $R_H^{(1)}$ is strongly temperature-dependent at low temperatures $T \leq t_b = 300$ K because of development of the “hot spots” in $\tau(k_y)$ and vanishes at high temperatures $T \geq t_b$. The different curves in Fig. 1 illustrate the sensitivity of $R_H^{(1)}$ to the choice of the phases φ and φ' in the transverse dispersion law (5), which determines the pattern of $\tau(k_y)$.

The temperature dependence of the total Hall coefficient $R_H(T)$, normalized to $R_H^{(0)}$, is shown in Fig. 2 for the 1/4-filled band with $t_a/t_b = 30$, which corresponds to $p_F v_F / t_b = 66.6$. We observe that all curves saturate at $R_H^{(0)}$ at high temperatures, and the curves (a) and (d) change sign at lower temperatures. The total Hall coefficient $R_H(T)$ may change sign only for

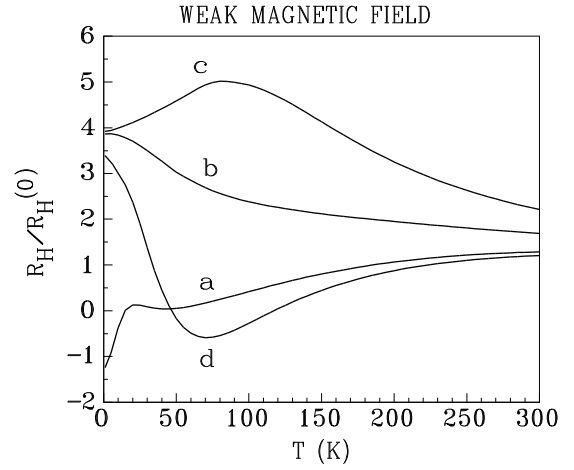


Figure 2: Temperature dependence of the Hall coefficient R_H , normalized to $R_H^{(0)}$, in the case $\omega_c \tau \ll 1$.

a sufficiently high anisotropy t_a/t_b , which makes $R_H^{(1)}$ comparable with $R_H^{(0)}$.

The above results apply in the limit of a weak magnetic field where $\omega_c \tau \ll 1$. The cyclotron frequency $\omega_c = 2\pi/t_0$ is defined in terms of the time t_0 it takes an electron to circle around the Fermi surface in the case of a closed orbit or traverse the Brillouin zone in the case of an open orbit. However, the magnetic fields 4–12 T used in the experiment [1] most likely correspond to the limit of a strong magnetic field: $\omega_c \tau \gg 1$. We study this case below.

A general analytical solution of Eq. (1) gives the following expression for the conductivity tensor [3]:

$$\sigma_{ij} = \frac{2ec}{(2\pi\hbar)^2 H} \frac{1}{1 - \exp[-(c/eH) \oint dp_t / v(p_t) \tau(p_t)]} \quad (17)$$

$$\times \oint dp_t \frac{v_i(p_t)}{v(p_t)} \oint_{p_t - P_t}^{p_t} dp'_t \frac{v_j(p'_t)}{v(p'_t)} \exp\left(\frac{-c}{eH} \int_{p'_t}^{p_t} \frac{dp''_t}{v(p''_t) \tau(p''_t)}\right),$$

where P_t is the period of integration over the tangential momentum p_t . Expanding Eq. (17) to the first power of $1/H$, we find the following expression for the Hall conductivity in the limit of a strong magnetic field:

$$\sigma_{xy} = -\frac{2ec}{(2\pi\hbar)^2 H} \oint dp_t / v(p_t) \tau(p_t) \quad (18)$$

$$\times \oint dp_y \oint_{p_t - P_t}^{p_t} dp'_x \int_{p'_t}^{p_t} \frac{dp''_t}{v(p''_t) \tau(p''_t)},$$

where dp_x and dp_y are the x and y projections of the differential dp_t . Taking the integral over dp'_x by parts, we find:

$$\sigma_{xy} = \frac{2ec}{(2\pi\hbar)^2 H} \quad (19)$$

$$\times \left(\oint dp_y p_x - \frac{1}{\oint dp_t/v\tau} \oint dp_y \oint_{p_t-P_t}^{p_t} \frac{dp'_t p'_x}{v(p'_t)\tau(p'_t)} \right).$$

For closed orbits, the integral $\oint dp_y$ in the second term in Eq. (19) vanishes, and the first term produces the familiar formula $\sigma_{xy} = enc/H$ [3]. However, the situation is the opposite for open orbits in Q1D conductors. In this case, the first term in Eq. (19) vanishes, if p_x is counted from the Fermi momentum p_F , whereas the second term does not vanish and gives the following expression:

$$\sigma_{xy} = -\frac{2ec}{\pi\hbar Hb} \frac{1}{\langle 1/v_x(k_y)\tau(k_y) \rangle_{k_y}} \left\langle \frac{p_x(k_y) - p_F}{v_x(k_y)\tau(k_y)} \right\rangle_{k_y}. \quad (20)$$

In this and the following equations (20)–(23), (25), and (26), the averaging over k_y is performed for the $+p_F$ sheet of the Fermi surface, and the result is doubled to account for the $-p_F$ sheet.

The Hall conductivity (20) vanishes if $\tau(k_y) = \tau$ and $v_x(k_y) = v_F$ are assumed to be constant over the Fermi surface. Thus, we need to take into account both terms in Eq. (8), which produce the following two contributions to the Hall conductivity when substituted into Eq. (20):

$$\sigma_{xy}^{(1)} = \frac{2ec}{\pi\hbar H v_F b} \frac{1}{\langle 1/\tau(k_y) \rangle_{k_y}} \left\langle \frac{\varepsilon_y(k_y)}{\tau(k_y)} \right\rangle_{k_y}, \quad (21)$$

$$\sigma_{xy}^{(2)} = -\beta \frac{2ec}{\pi\hbar H p_F v_F^2 b} \frac{1}{\langle 1/\tau(k_y) \rangle_{k_y}} \left\langle \frac{\varepsilon_y^2(k_y)}{\tau(k_y)} \right\rangle_{k_y}. \quad (22)$$

The longitudinal conductivity is given by the zeroth-order term of expansion of Eq. (19) in powers of $1/H$:

$$\sigma_{xx} = \frac{2e^2}{\pi\hbar b \langle 1/v_x(k_y)\tau(k_y) \rangle_{k_y}} \approx \frac{2e^2 v_F}{\pi\hbar b \langle 1/\tau(k_y) \rangle_{k_y}}. \quad (23)$$

The transverse conductivity is given by the second-order term of expansion of Eq. (19) in powers of $1/H$. The term originating from expansion of the first line in Eq. (17) is similar to Eq. (19) with the integral over p_y replaced by an integral over p_x . This term vanishes. Another term, originating from expansion of the second line in Eq. (17), has the following form:

$$\sigma_{yy} = \frac{c^2}{(2\pi\hbar)^2 H^2} \oint dp_t/v(p_t)\tau(p_t) \times \oint dp_x \oint_{p_t-P_t}^{p_t} dp'_x \left(\int_{p'_t}^{p_t} \frac{dp''_t}{v(p''_t)\tau(p''_t)} \right)^2. \quad (24)$$

Taking the integrals over p'_x and p_x by parts, we find:

$$\sigma_{yy} = \frac{2c^2}{\pi\hbar H^2 b \langle 1/v_x(k_y)\tau(k_y) \rangle_{k_y}} \times \left(\left\langle \frac{(p_x - p_F)^2}{v_x \tau} \right\rangle_{k_y} \left\langle \frac{1}{v_x \tau} \right\rangle_{k_y} - \left\langle \frac{p_x - p_F}{v_x \tau} \right\rangle_{k_y}^2 \right). \quad (25)$$

The second term in the brackets in Eq. (25) vanishes when $\tau(k_y) = \text{const}$ and $v_x(k_y) = \text{const}$, whereas the first term does not. Thus, the second term is much smaller than the first term and can be neglected. Besides, the second term exactly cancels with σ_{xy}^2 (20) in the combination $\sigma_{xx}\sigma_{yy} + \sigma_{xy}^2$ that appears in $R_H = \sigma_{xy}/H(\sigma_{xx}\sigma_{yy} + \sigma_{xy}^2)$. Thus, in Q1D conductors with open electron orbits, $\sigma_{xy}^2 \ll \sigma_{xx}\sigma_{yy}$ even in the limit $\omega_c\tau \gg 1$, unlike in metals with closed electron orbits. Neglecting the second term in Eq. (25), we find:

$$\sigma_{yy} \approx \frac{2c^2}{\pi\hbar H^2 v_F^3 b} \left\langle \frac{\varepsilon_y^2(k_y)}{\tau(k_y)} \right\rangle_{k_y}. \quad (26)$$

Using Eqs. (21), (22), (23), and (26), we find two contributions to the Hall coefficient:

$$R_H = R_H^{(1)} + R_H^{(2)} = \frac{\sigma_{xy}^{(1)}}{H\sigma_{xx}\sigma_{yy}} + \frac{\sigma_{xy}^{(2)}}{H\sigma_{xx}\sigma_{yy}}. \quad (27)$$

In Fig. 3, we show the temperature dependence $R_H(T)$ in the case $\omega_c\tau \gg 1$ calculated for the same model with $t_a/t_b = 30$ as in Fig. 2. At high temperatures, where $\tau(k_y)$ does not depend on k_y , $R_H^{(1)}$ vanishes, whereas $R_H^{(2)}$ saturates at $R_H^{(0)}$ given by Eq. (16). At lower temperatures, where the “hot spots” in $\tau(k_y)$ develop, $R_H^{(1)}$ and, consequently, R_H become strongly temperature-dependent. However, $R_H^{(1)}$ vanishes at $T = 0$, and R_H returns to the value $R_H^{(0)}$. This is related to the peculiar expression for the relaxation rate due to umklapp scattering along the chains at $T = 0$ in our model [2]:

$$1/\tau(k_y) \propto T^2 \int dk_y^{(1)} dk_y^{(2)} dk_y^{(3)} \delta(k_y + k_y^{(1)} - k_y^{(2)} - k_y^{(3)}) \times [\varepsilon_y^+(k_y) + \varepsilon_y^+(k_y^{(1)}) + \varepsilon_y^-(k_y^{(2)}) + \varepsilon_y^-(k_y^{(3)})] \quad (28)$$

where $\varepsilon_y^+(k_y)$ and $\varepsilon_y^-(k_y)$ are the transverse dispersion laws (5) for the $\pm p_F$ sheets. The average over k_y that appears in Eq. (21) vanishes, because it can be written as an average of the argument of the delta-function:

$$\langle \varepsilon_y(k_y)/\tau(k_y) \rangle_{k_y} \propto (1/4) \int dk_y dk_y^{(1)} dk_y^{(2)} dk_y^{(3)} \times \delta(k_y + k_y^{(1)} - k_y^{(2)} - k_y^{(3)}) \times [\varepsilon_y^+(k_y) + \varepsilon_y^+(k_y^{(1)}) + \varepsilon_y^-(k_y^{(2)}) + \varepsilon_y^-(k_y^{(3)})] \times \delta[\varepsilon_y^+(k_y) + \varepsilon_y^+(k_y^{(1)}) + \varepsilon_y^-(k_y^{(2)}) + \varepsilon_y^-(k_y^{(3)})] = 0 \quad (29)$$

In conclusion, we have developed a systematic theory of the Hall effect in Q1D conductors for the model where the electron relaxation time $\tau(k_y)$ varies over the Fermi surface. We have studied the cases of both weak ($\omega_c\tau \ll 1$) and strong ($\omega_c\tau \gg 1$) magnetic fields. At high temperatures, the Hall coefficient saturates at the value $-\beta/ecn$, where the dimensionless coefficient β is determined by the second derivative $\partial^2 \varepsilon_x / \partial p_x^2$ of the longitudinal dispersion law of electrons (see Eqs. (6)

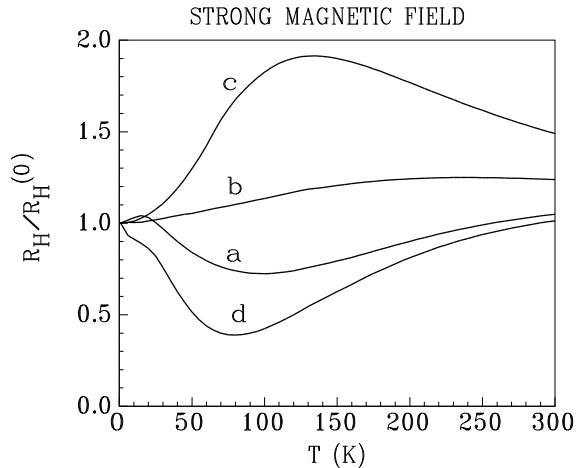


Figure 3: Temperature dependence of the Hall coefficient R_H , normalized to $R_H^{(0)}$, in the case $\omega_c\tau \gg 1$.

and (7)). At low temperatures, where a strong variation of the relaxation rate over the Fermi surface develops in the form of “hot spots”, R_H becomes temperature-dependent and may change sign for a particular choice of the transverse dispersion law parameters. In our model, the sign changes in a weak, but not in a strong magnetic field.

We need to add two caveats to our study. First, the electron relaxation rate in this paper and in Ref. [2] was calculated for umklapp scattering along the chains. While this scattering is believed to be relevant for calculating σ_{xx} , it is not clear whether it is also relevant for calculating of σ_{xy} and σ_{yy} . On the other hand, our Eq. (1) requires to use the same relaxation time for all transport coefficients. Because of this uncertainty, our numerical calculations of R_H should be considered only as a qualitative illustration. Second, the sign change of $R_H(T)$ was experimentally found in Ref. [1] in the regime where the temperature dependence of resistivity changes slope from metallic to insulating, which may be due to opening of a charge pseudogap [7]. Our conventional, Fermi-liquid treatment of transport coefficients may not apply in this complicated situation.

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